Definition of a scoring parameter to identify low-dimensional materials components

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I. INTRODUCTION

Low-dimensional materials with one or more characteristic lengths of the materials limited to the atomic scale have received significant attention recently. Since the discovery of graphene the world has seen intense research in 2D materials both in their bulk form, as individual layers a few atoms thick, and through stacking of 2D layers into heterostructures. The identification of low-dimensional compounds is therefore of key interest. Here, we perform a geometric analysis of material structures, demonstrating a strong clustering of materials depending on their dimensionalities. Based on the geometric analysis, we propose a simple scoring parameter to identify materials of a particular dimension or of mixed dimensionality. The method identifies spatially connected components of the materials and gives a measure of the degree of “1D-ness,” “2D-ness,” etc., for each component. The scoring parameter is applied to the Inorganic Crystal Structure Database and the Crystallography Open Database, ranking the materials according to their degree of dimensionality. In the case of 2D materials the scoring parameter is seen to clearly separate 2D from non-2D materials and the parameter correlates well with the bonding strength in the layered materials. About 3000 materials are identified as one-dimensional, while more than 9000 are mixed-dimensionality materials containing a molecular (0D) component. The charge states of the components in selected highly ranked materials have either zero charge, corresponding to weak interactions, or integer charge, indicating ionic bonding.
Here, $d_{ij}$ is the distance between atoms $i$ and $j$, $r_{ic}^i$ and $r_{ic}^j$ are the corresponding covalent radii [13], and $k$ is a variable to be investigated. The latter choice is motivated by the strong dependence of the classification of the dimensionality of a material upon the $k$ value; as illustrated for the boron-nitride structure in Fig. 1(a), too small a $k$ value will underestimate the dimensionality, whereas too large a $k$ value will overestimate it. Rather than attempt to identify a good value of $k$, we observe that, for any given structure, there exists a finite number of relevant $k$ intervals to investigate.

We start by considering the set of interatomic distances in a material, sorted by increasing $k$ value; as illustrated for the boron-nitride structure in Fig. 1(a), too small a $k$ value will underestimate the dimensionality, whereas too large a $k$ value will overestimate it. Rather than attempt to identify a good value of $k$, we observe that, for any given structure, there exists a finite number of relevant $k$ intervals to investigate.

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The position of the cluster is intuitive from a bonding perspective. First, if the bonding model and covalent radii exactly described the actual bond lengths, the cluster would lie on the line $k_1 = 1$; the variability in the interval starting points results from the simplicity of the ball-and-stick bonding model. Second, since low-dimensional components are geometrically separated, we should expect a correspondingly wide $k$ interval; it can be seen that easily exfoliable structures such as graphite, boron nitride, and molybdenum disulfide have wide $k$ intervals.

We propose a scoring parameter which distills the above observations of the $k$-interval plots into a single number:

$$s(k_1, k_2) = f(k_2) - f(k_1),$$

where

$$f(x) = \frac{c \times \max(0, x - 1)^2}{1 + c \times \max(0, x - 1)^2}.$$  

Here, $c$ is a constant which determines the scale at which a bond is broken. We use $c = 1/0.15^2$, which is chosen so that $s(1, 1.15) = s(1.15, \infty) = 0.5$; slightly different values of the parameter will give similar results. Figure 3 illustrates how a $k$ interval is transformed into a score.

The interval width increases the score, but with diminishing returns as $k_1$ increases above 1. This avoids the $[k, \infty)$ interval dominating unless $k_1$ is close to 1, in which case the structure is indeed 3D. Furthermore, $k$ values below 1 are effectively set to 1; this avoids erroneous low-dimensional classifications when $[k_1, k_2] \approx [0, 1]$. In structures with low-dimensional components, the scoring parameter rewards large intercomponent distances. A further convenient property of the scoring scheme is that the interval scores sum to 1. We have found that the best results are achieved by merging $k$ intervals with the same types of dimensionality [e.g., intervals of the same color in Fig. 1(b)].
The principal motivation of the scoring scheme is to identify the intuitively correct dimensionality classification, by determining whether a $k$ interval lies within a cluster of the type shown in Figs. 2(a) and 2(b). Using the scoring parameter, the structures are colored in Figs. 2(c) and 2(d) according to their dimensionality classification.

The scoring scheme is demonstrated for three structures in Fig. 4. The first material, Al$_2$O$_3$, is clearly a bulk crystalline structure. If a single $k$-value threshold at $k \approx 1$ were used, however, it would result in a misclassification as a layered structure. Similarly, the AuTe$_2$ has an ambiguous classification, lying close to the contour $s_2(k_1, k_2) = 0.5$. In this case the dimensionality classification is sensitive to small changes in the functional form or the parameters of the scoring function. Then, the useful information contained in the scores is not in their exact values, but rather that $s_2$ and $s_3$ are approximately equal in value; this can be interpreted as a layered structure with a very small interlayer spacing.

It should be emphasized that the scoring is exclusively based on interatomic distances and atomic sizes, and that it simply assumes that longer bonds tend to be weaker than shorter ones. The physical characters of the bonds, i.e., whether they can be considered covalent, ionic, or of dispersion type, are not revealed. Nonetheless, the coarse treatment of bond lengths is justified by the cluster separation in Fig. 2(b). We will show that the scoring scheme allows for identification of interesting materials, whose properties can then be investigated experimentally or using electronic structure methods.

The scaled bonding criterion described in Eq. (1) is the same one employed by Ashton et al. [6] in their study of layered materials. An additive bonding criterion of the form $d_{ij} < r_i + r_j + \Delta$ is used by Mounet et al. [5] and Cheon et al. [14], using van der Waals radii and elemental radii, respectively. In these works, the material dimension is determined by sampling a range of parameter values (either $k$ or $\Delta$) in a fixed interval, which does not easily permit the construction of a scoring parameter. Cluster dimensionalities...
Sodium-ion batteries. McKinney et al. [20] have extended this to include the analysis of the packing fraction [15,16], identifying layered materials with components of higher dimensionality. These correspond to materials with one or more 0D components in combination with materials of different dimensionality. In particular there are 9459 materials which have one or more 0D components in combination with components of higher dimensionality. These correspond to molecules or molecular ions embedded in the higher-dimensional network. Only a few materials combine 1D, 2D, and 3D components. We find 15 materials combining 1D and 2D. There are 22 materials which combine 1D and 3D components. Three of them are shown in Fig. 5.

We have made every effort to remove inconsistent structures from the database, an automated filtering is not sufficient given the many different types of errors and partial structures present in the ICSD and COD. The numbers presented here should therefore be taken as only approximate.

A database containing the calculated scoring parameters for all dimensionalities for all compounds in the ICSD and COD is available at the Computational Materials Repository [23].

D. Physical significance of the scoring parameter

Due to the well-defined identification of the 2D materials, the scoring scheme also serves as a simple predictor of exfoliability. Mounet et al. [5] have calculated the exfoliation energy (i.e., the binding energy between layers) of 1535 layered materials, and they suggest an energy of 35 meV/Å² as the
threshold for “easily exfoliable” materials. They furthermore highlight 11 materials, which they denote as “well-known” 2D materials.

In Fig. 6 we show the calculated exfoliation energies versus the scoring parameter $s_2$. There is a clear correlation between the scoring parameter and the exfoliation energy with essentially all of the high-scoring materials (say $s_2 > 0.7$) having an exfoliation energy below the threshold. The separation of materials of different dimensionality is also clearly seen here by the low density of points in the region $s_2 \approx 0.3 – 0.5$. The 11 well-known 2D materials are also shown in the figure. All of them, except Bi$_2$Te$_3$, have high-scoring values with $s_2 > 0.7$. Despite its small interlayer distance, Bi$_2$Te$_3$ is nonetheless classified as a 2D material, since $s_2$ is larger than its other scores.

It should be noted that although the exfoliation energy is a highly relevant quantity for the exfoliation process, it is not clear whether an absolute threshold in energy is the best indicator of exfoliability. The exfoliation process involves breaking the bonds between the layers keeping the bonds within the layers intact, so the exfoliation energy should be seen relative to the intralayer bond strengths. While the scoring parameter proposed here does not explicitly involve the energetics, the high-scoring materials have a clear separation between the intra- and intercomponent bond lengths, which can be expected to be a characteristic of easily exfoliable materials.

**E. Ranking of low-dimensional materials**

In addition to dimensionality classification, the scoring parameter defines an order on materials. We have identified the ten materials in the ICSD and COD with the highest 2D scores, shown in Table III. Widely studied layered structures such as graphene, boron nitride, and magnesium chloride are highly ranked. Some of the remaining structures have much larger unit cells, but are nonetheless clearly van der Waals
TABLE I. Number of structures remaining after each stage of filtering, performed in the order shown. “Defective” structures encompass incorrect CIF files and theoretical structures, and manually identified entries such as misfit compounds, surface structures, and superstructures. Where duplicate structures are found across the two databases, the COD structure is kept.

<table>
<thead>
<tr>
<th>Dimensions</th>
<th>COD Removed</th>
<th>COD Remaining</th>
<th>ICSD Removed</th>
<th>ICSD Remaining</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>400731</td>
<td>184754</td>
<td></td>
<td></td>
</tr>
<tr>
<td>&gt;200 atoms</td>
<td>185329</td>
<td>215402</td>
<td>7474</td>
<td>177280</td>
</tr>
<tr>
<td>Partial occupancy</td>
<td>49015</td>
<td>166387</td>
<td>75659</td>
<td>101621</td>
</tr>
<tr>
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<td>1470</td>
<td>164917</td>
<td>7184</td>
<td>94437</td>
</tr>
<tr>
<td>Defective</td>
<td>10219</td>
<td>154698</td>
<td>5703</td>
<td>88734</td>
</tr>
<tr>
<td>Duplicates</td>
<td>15646</td>
<td>139052</td>
<td>60019</td>
<td>28715</td>
</tr>
<tr>
<td>Total remaining</td>
<td>167767</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

TABLE II. Number of entries of each dimensionality type found in the ICSD and COD. In the diagonal the number of materials with a single dimension are shown while the off-diagonal entries indicate materials with components of two different dimensionalities. In addition to the single- and bidimensional materials counted here, we have found 16 tridimensional structures with 0D, 1D, and 2D components.

<table>
<thead>
<tr>
<th>Dimensions</th>
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<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
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<td>0</td>
<td>105199</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td></td>
</tr>
<tr>
<td>2</td>
<td>2946</td>
<td>15</td>
<td>4623</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>3010</td>
<td>22</td>
<td>0</td>
<td>45148</td>
</tr>
</tbody>
</table>
FIG. 6. Binding energies ($E_b$) vs 2D scores for 1535 layered materials identified by Mounet et al. [5], colored according to dimensionality classification. Structures with $E_b < 35$ meV/Å$^2$ are classified by Mounet et al. as easily exfoliable. For clarity, 44 01D, five 03D, and four 012D structures are not shown here.

As mentioned in the introduction several computational 2D materials databases are presently under construction while 1D materials and materials of mixed dimensionality have received much less attention. The present approach allows for simple identification of existing 1D or mixed-dimensional materials, which can form templates that can be used to construct larger computational databases for materials of a given dimensionality.

### IV. METHODS

#### A. Component dimensionality

A material will in general consist of several clusters of bonded atoms. Such clusters we term the *components* of the material. The components may have different dimensionalities and they should therefore be investigated separately.

Our definition of material dimensionality of a component is as follows: select an atom in the component, with atomic coordinates $x_1$. Let $X = \{x_1, x_2, x_3, \ldots, x_i\}$ denote the set of atoms to which the first atom is bonded, and which have the same fractional coordinates but in different unit cells, i.e., $x_i = x_1 + C^T h_i$, where $C$ is the unit cell description and $h_i$ is an integer vector. Then, the component dimensionality is the rank of the subspace spanned by $X$:

$$\dim(X) = \text{rank}\left(\{x_2 - x_1, x_3 - x_1, \ldots, x_i - x_1\}\right). \quad (4)$$

This definition (illustrated in Fig. 8) accommodates both corrugation and thickness. While $X$ is an infinite set for all but 0D components, $\dim(X)$ can be determined in a finite number of steps by exploiting the periodicity of a material.

As described above, determination of the dimension of a material requires an analysis of its constituent bonded clusters, or *components*. To find the dimension of a component, the rank determination algorithm (RDA) of Mounet et al. [5] uses a supercell of fixed size with open boundary conditions. If the supercell is too small, the number of components might be overestimated.

Conversely, the topological scaling algorithm (TSA) of Ashton et al. [6] uses periodic unit cells, which can underestimate the number of components by forming improper connections between them. By improper connections, we mean components which are disconnected in the infinite crystal but are connected due to the periodic cell chosen.

### TABLE III. Top ten “most 2D” materials in the ICSD and COD, as ordered by the interval scoring method in Eq. (2).

<table>
<thead>
<tr>
<th>Source</th>
<th>ID</th>
<th>Compound</th>
<th>$s$</th>
<th>$k_1$</th>
<th>$k_2$</th>
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</thead>
<tbody>
<tr>
<td>COD</td>
<td>1000410</td>
<td>TiAlF$_4$</td>
<td>0.987</td>
<td>0.992</td>
<td>2.330</td>
</tr>
<tr>
<td>COD</td>
<td>9000046</td>
<td>C</td>
<td>0.986</td>
<td>0.933</td>
<td>2.251</td>
</tr>
<tr>
<td>ICSD</td>
<td>27987</td>
<td>BN</td>
<td>0.984</td>
<td>0.935</td>
<td>2.170</td>
</tr>
<tr>
<td>ICSD</td>
<td>248235</td>
<td>C$_2$N$_4$</td>
<td>0.983</td>
<td>0.910</td>
<td>2.155</td>
</tr>
<tr>
<td>ICSD</td>
<td>187384</td>
<td>Rb(Au(CF$_3$SO$_3$)$_4$)</td>
<td>0.982</td>
<td>1.010</td>
<td>2.313</td>
</tr>
<tr>
<td>ICSD</td>
<td>163023</td>
<td>Sr(CF$_3$SO$_2$)$_2$N$_2$</td>
<td>0.981</td>
<td>1.014</td>
<td>2.516</td>
</tr>
<tr>
<td>COD</td>
<td>1525422</td>
<td>K$_2$Mn(CN)$_6$</td>
<td>0.980</td>
<td>0.935</td>
<td>2.039</td>
</tr>
<tr>
<td>COD</td>
<td>1534338</td>
<td>MgCl$_2$</td>
<td>0.977</td>
<td>0.959</td>
<td>1.976</td>
</tr>
<tr>
<td>ICSD</td>
<td>161278</td>
<td>B$<em>3$C$</em>{10}$N$_3$</td>
<td>0.977</td>
<td>0.965</td>
<td>1.968</td>
</tr>
<tr>
<td>COD</td>
<td>2242431</td>
<td>Cs(N(SO$_2$CF$_3$)$_2$)</td>
<td>0.977</td>
<td>0.965</td>
<td>1.968</td>
</tr>
</tbody>
</table>

### TABLE IV. Top ten “most 1D” hydrogen-free materials in the ICSD and COD, as ordered by the interval scoring method in Eq. (2).

<table>
<thead>
<tr>
<th>Source</th>
<th>ID</th>
<th>Compound</th>
<th>$s$</th>
<th>$k_1$</th>
<th>$k_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD</td>
<td>4344111</td>
<td>NPF$_2$</td>
<td>0.984</td>
<td>0.889</td>
<td>2.184</td>
</tr>
<tr>
<td>COD</td>
<td>9010982</td>
<td>SO$_3$</td>
<td>0.983</td>
<td>0.954</td>
<td>2.133</td>
</tr>
<tr>
<td>ICSD</td>
<td>72577</td>
<td>Ru(CO)$_4$</td>
<td>0.982</td>
<td>0.979</td>
<td>2.095</td>
</tr>
<tr>
<td>ICSD</td>
<td>47183</td>
<td>Cu(CO)$_4$</td>
<td>0.980</td>
<td>0.926</td>
<td>2.044</td>
</tr>
<tr>
<td>ICSD</td>
<td>291211</td>
<td>Si$_2$</td>
<td>0.967</td>
<td>0.986</td>
<td>1.815</td>
</tr>
<tr>
<td>ICSD</td>
<td>22090</td>
<td>RuCl$_3$</td>
<td>0.967</td>
<td>0.940</td>
<td>1.812</td>
</tr>
<tr>
<td>ICSD</td>
<td>415951</td>
<td>V(AlCl$_4$)$_2$</td>
<td>0.965</td>
<td>0.990</td>
<td>1.784</td>
</tr>
<tr>
<td>ICSD</td>
<td>78777</td>
<td>CrF$_3$</td>
<td>0.964</td>
<td>0.985</td>
<td>1.780</td>
</tr>
<tr>
<td>ICSD</td>
<td>428185</td>
<td>AlPS$_4$</td>
<td>0.964</td>
<td>0.995</td>
<td>1.771</td>
</tr>
<tr>
<td>ICSD</td>
<td>419661</td>
<td>CrF$_3$</td>
<td>0.962</td>
<td>0.996</td>
<td>1.751</td>
</tr>
</tbody>
</table>
The problem of improper connections is illustrated with a contrived example in Fig. 9, which shows the side view of a selection of periodic helical structures. We define an \( n \)-helix as a structure which has \( n \) components, whose \( j \)th component has coordinates given by

\[
x_j = \sin \frac{2\pi (t + j)}{n}, \quad y_j = \cos \frac{2\pi (t + j)}{n}.
\]

The number of components is dependent on the size (along the \( t \) axis) of the periodic cell. In the formulation given in Eq. (5) any integer is a valid cell length.

Figure 9 shows how the number of components changes with varying cell periodicity. In general, the number of components for an \( n \)-helix with periodicity \( m \) is given by gcd \((n, m \mod n)\). In order to avoid improper connections between components, a periodic cell of size \( n \) is needed. This is further complicated for cells containing multiple \( n \)-helices of different sizes. In this case, the correct size of the periodic cell is given by \( \text{lcm}(n_1, n_2, n_3, \ldots) \), where \( n_i \) denotes the number of components in the \( i \)th \( n \)-helix. For example, a structure containing a 5-helix, a 6-helix, and a 7-helix requires a periodic cell of length 210. This cell is so large that it is unlikely that it would be tested using the existing methods.

While the example described here is contrived, self-penetrating helical networks have been assembled experimentally \([32,33]\). Furthermore, the problem illustrated has practical consequences: an incorrect periodic cell (such as the use of a primitive unit cell) causes the interpenetrating polymer networks shown in Fig. 5 to be misclassified as 2D materials.

**B. Algorithm**

Component dimensionalities can be identified using a modified breadth-first-search (BFS) algorithm, shown in Algorithm 1. In standard BFS, the search terminates when all nodes have been visited. Here, we terminate the search when the rank of the subspace spanned by a component (i.e., the dimensionality) can no longer increase. The rank of a set of points is defined as

\[
\text{rank}(\mathbf{v}) = \begin{cases} 
-1, & \text{if } \mathbf{v} = \emptyset, \\
\text{rank}_M(\mathbf{v} - \mathbf{v}_i), & \text{otherwise},
\end{cases}
\]

where \( \text{rank}_M \) denotes the standard matrix rank.

In this algorithm, components in the aperiodic primitive unit cell are the graph vertices, and connections between components (across unit cell boundaries) are graph edges. We note that, by definition, no edges exist between components within the same cell.

The input to the algorithm (line 1) is a set of graph edges (\( E \)) and a component (\( c \)) whose dimension we wish to determine. We maintain a set of visited or seen vertices (line 2) and a set of visited vertices for each of the \( n \) components in the aperiodic primitive unit cell (line 3). A vertex queue is maintained whose elements consist of a component index and cell coordinates. The queue is initialized with the component \( c \) in the cell with coordinates \( O = [0, 0, 0] \) (line 4). The algorithm runs until the queue is empty (line 5). The first element in the queue is extracted and removed (lines 6 and 7). If the element has already been visited it is skipped (line 8); otherwise it is added to the set of visited elements (line 11). If the addition of the vertex serves to increase the rank of the set of visited vertices (line 12), it is added to the set (line 13).

New vertices in adjacent cells are generated from the edge list. For a component \( i \), the edge list \( E_i \) (line 15) specifies the neighboring components (\( j \)) and the cell offset (\( \Delta \)), from which the coordinates of the neighboring cell can be calculated (line 16). If the neighbor element has already been visited it is either skipped (line 17), or added to the queue (line 21) if it serves to increase the rank of the set of visited vertices (line 20). When the queue is empty, the rank of the component is returned (line 25).

**C. Interval identification**

The purpose of the modified method (described in Algorithm 2) is to identify intervals in \( k \) in which the dimensionality classification is constant.
The infinite structure contains 8 components. Any number of repetitions, connections between components. Figure 9. Improper connections between components in \( n \)-helix structures, here for \( n = 8 \). The number of components is denoted by \( n_c \). The infinite structure contains 8 components. Any number of repetitions, \( m \geq 1 \), of the cell for which \( m \) mod 8 \( \neq 0 \) results in improper connections between components.

The algorithms developed here are included in the ASE [27] library.

Algorithm 1. Pseudocode for calculating component dimensionality.

```plaintext
1: procedure CALCULATEDIMENSIONALITY(E, c)
2: s := ∅
3: v := \{∅ \forall i \in 1 \ldots n\}
4: Q := \{(c, 0)\}
5: while Q ≠ ∅ do
6: (i, p) := Q
7: Q := Q \ Q
8: if (i, p) ∈ s then
9: \ Continue
10: end if
11: s := s ∪ \{(i, p)\}
12: if rank \((v_i ∪ \{p\}) > rank \((v_i)\) then
13: v_i := v_i ∪ \{p\}
14: end if
15: for \((j, \Delta) \in E\) do
16: \( \tilde{q} := \tilde{p} + \Delta \)
17: if \(\{(j, \tilde{q})\} \in s\) then
18: \ Continue
19: end if
20: if rank \((v_j ∪ \{\tilde{q}\}) > rank \((v_j)\) then
21: Q := Q ∪ \{j, \tilde{q}\}
22: end if
23: end for
24: end while
25: return rank \((v_c)\)
26: end procedure
```

Algorithm 2. Pseudocode for finding all dimensionality intervals.

```plaintext
1: procedure FINDINTERVALS (E)
2: \( k_{prev} := 0 \)
3: \( h_{prev} := [n_{atoms}, 0, 0, 0] \)
4: \( R := ∅ \)
5: for \((k, i, j) \in E\) do
6: Add edge between vertices \(i\) and \(j\)
7: Identify connected components
8: Update \(h\)
9: if \(h \neq h_{prev}\) then
10: \( R := R ∪ (k_{prev}, k, h_{prev})\)
11: end if
12: if \(h = [0, 0, 0, 1]\) then
13: return \(R ∪ ((k, ∞, h))\)
14: end if
15: \( k_{prev} := k \)
16: \( h_{prev} := h \)
17: end for
18: end procedure
```
ACKNOWLEDGMENTS

The authors thank Nicolas Mounet and Nicola Marzari for kindly providing data for the layered compounds identified by Mounet et al. [5], FIZ Karlsruhe–Leibniz Institute for Information Infrastructure for providing CIF files of all entries in the ICSD, and anonymous referees for comments which improved the manuscript. This work was supported by Grant No. 7026-00126B from the Danish Council for Independent Research and by the VILLUM Center for Science of Sustainable Fuels and Chemicals which is funded by the VILLUM Fonden research grant (9455).


